Repulsive shield between polar molecules

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(Dated: May 5, 2008)

We propose and analyze a technique that allows to suppress inelastic collisions and simultaneously enhance elastic interactions between cold polar molecules. The main idea is to cancel the leading dipole-dipole interaction with a suitable combination of static electric and microwave fields in such a way that the remaining van-der-Waals-type potential forms a three-dimensional repulsive shield. We analyze the elastic and inelastic scattering cross sections relevant for evaporative cooling of polar molecules and discuss the prospect for the creation of crystalline structures.

PACS numbers: 34.50.Rk, 37.10.Mn, 64.70.dg

It is well known that an attractive van der Waals interaction dominates the microscopic interaction potential between ground state atoms and molecules at short distances [1]. As a consequence, ultracold atomic and molecular gases are prone to the formation of deeply bound molecular states via inelastic collisions. These inelastic collisions limit the lifetime of strongly interacting ultracold gases [1, 2]. Controlling inelastic collisions is therefore crucial for the creation of novel strongly correlated quantum degenerate systems, such as polar molecules. In this Letter, we demonstrate the possibility to engineer a three-dimensional repulsive interaction between polar molecules, which allows for the suppression of inelastic collisions, while simultaneously enhancing elastic collisions. This technique may open up a way towards efficient evaporative cooling and the creation of novel long-lived quantum degenerate gases of polar molecules.

A special property of polar molecules prepared into the lowest rotational and vibrational state is a permanent electric dipole moment d, which gives rise to tunable dipole-dipole interactions and offers a large potential for the creation of strongly correlated quantum phases [3, 4, 5, 6]. Two routes are currently explored for the experimental realization of quantum degenerate polar molecules: (i) trapping and cooling of preexisting molecules [7, 8, 9, 10, 11, 12] and (ii) synthesizing polar molecules from a cold mixture of atomic gases [13, 14, 15, 16, 17, 18, 19]. While scattering properties of heteronuclear molecules with dipole-dipole interactions are currently theoretically explored [20, 21, 22], it is expected that inelastic collisions strongly increase for polar molecules compared to atomic systems due to the opening of additional decay channels.

The main idea of our approach is to cancel the leading dipole-dipole interaction with a suitable combination of a static electric field and a continuous-wave microwave field: the former induces a dipole moment d_z, while the latter drives an additional dipole moment $d_\perp$ rotating with frequency ω of the microwave field, see Fig. 1(b). In analogy with magic-angle techniques in NMR [23], the time-averaged interaction of the rotating dipole moment shows a negative sign allowing for a cancelation of the total dipole-dipole interaction. The remaining interaction is tunable in strength with a repulsive van der Waals behavior $V_{\rm eff} \sim (d^4/\hbar\Delta)/r^6$, where Δ is the detuning of the microwave field and r is the intermolecular separation. The three-dimensional shield described here is thus purely repulsive. This is in contrast to the "blue shield" discussed in the context of atomic gases, which is attractive at large distances [1]. We find that the efficiency of the shield is determined by a single dimensionless parameter $\gamma = d^2 m/\hbar^2 r_{\rm B}$ with $r_{\rm B} = (d^2/B)^{1/3}$, B the rotational energy, and m the mass of the molecule. Under optimal conditions, for large values of γ, inealstic collisions can be quenched for temperatures $T \lesssim 0.01B$.

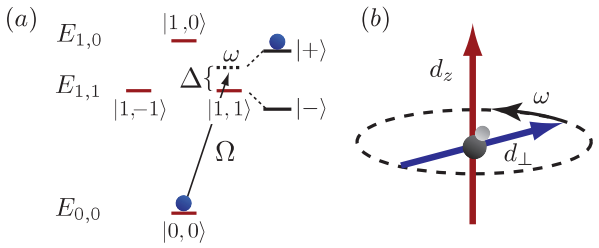

FIG. 1: (a) Energy levels of $H^{(i)}_{\rm rot}$: the arrow indicates the microwave field. (b) Sketch for the cancelation of the dipole-dipole interaction: d_z denotes the dipole moment induced by the static electric field, while the dipole moment $d_\perp$ is rotating in an orthogonal plane due to the microwave coupling.

To describe the shield quantitatively, we consider two polar molecules prepared into the vibrational ground state. The interaction at large intermolecular distances is dominated by the dipole-dipole interaction, yielding the

Hamiltonian

$$H = \frac{\mathbf{P}^2}{4m} + \frac{\mathbf{p}^2}{m} + \frac{\mathbf{d}_1\mathbf{d}_2 - 3(\mathbf{d}_1\hat{\mathbf{r}})(\mathbf{d}_2\hat{\mathbf{r}})}{r^3} + \sum_{i=1}^{2} H_{\rm rot}^{(i)}, \quad (1)$$

where we have introduced the center of mass $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$ and the relative coordinate $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$, with the corresponding momenta $\mathbf{P}$ and $\mathbf{p}$, respectively, while $r = |\mathbf{r}|$ and $\hat{\mathbf{r}} = \mathbf{r}/r$. Here, we are interested in polar molecules in the $^1\Sigma$ electronic ground state, and the Hamiltonian for the internal structure takes the form [24]

$$H_{\rm rot}^{(i)} = B\,\mathbf{J}_i^2 - \mathbf{d}_i \cdot \mathbf{E}_{\rm dc} - \mathbf{d}_i \cdot \mathbf{E}_{\rm ac}(t) \quad (2)$$

with the dipole operator $\mathbf{d}_i$ and the permanent dipole moment d. The first term describes a rigid rotor accounting for rotational structure with the rotational energy B, while the last two terms describe the coupling to an external static electric field $\mathbf{E}_{\rm dc}$ and microwave field $\mathbf{E}_{\rm ac}$. While additional interactions with the nuclear spins are in general small and can be ignored, the analysis presented here remains valid for polar molecules with an electronic spin in a strong magnetic field with the Zeeman splitting larger than the energy scales of the shield and the spin-rotation coupling; such a situation naturally appears for some polar molecules in magnetic traps.

We choose to apply a static electric field $\mathbf{E}_{\rm dc} = E_{\rm dc}\mathbf{e}_z$ along the z-axis. For each molecule, a suitable basis set for the internal states is given by the eigenstates of the rotor Hamiltonian in the external static field. These states and the corresponding energies depend on the dimensionless parameter $dE_{\rm dc}/B$ and are denoted by $|j, m_z\rangle_i$ and E_{j,m_z}, respectively, with m_z the angular momentum along the z-axis and j denoting the different energy manifolds, see Fig. 1(a). In addition, we apply a circularly (σ_+) polarized microwave field $\mathbf{E}_{\rm ac}(t)$ propagating along the z-axis and coupling dominantly the ground state $|0,0\rangle_i$ with the first excited state $|1,1\rangle_i$. The microwave field is characterized by the detuning $\Delta = \omega - (E_{1,1} - E_{0,0})/\hbar$ and Rabi frequency $\Omega = E_{\rm ac}d_c/\hbar$ with the dipole coupling $d_c = |\langle 0,0|\mathbf{d}_i|1,1\rangle|$. The leading effect of the microwave field on the internal structure of each molecule is to mix the ground state $|0,0\rangle$ with the excited state $|1,1\rangle$. We are interested in the regime with $\Delta, \Omega \ll B/\hbar$ and $dE_{\rm dc} < 2B$, where the rotating wave approximation is valid. In the rotating frame, these dressed states then take the form $|+\rangle = \alpha|0,0\rangle + \beta|1,1\rangle$ and $|-\rangle = \beta|0,0\rangle - \alpha|1,1\rangle$ with the energy splitting $\Delta E = \hbar\sqrt{\Delta^2 + 4\Omega^2}$ and $\alpha = -A/\sqrt{A^2+\Omega^2}$, $\beta = \Omega/\sqrt{A^2+\Omega^2}$, and $A = (\Delta + \sqrt{\Delta^2+4\Omega^2})/2$. Throughout this letter, we are interested in a shield with a high barrier, which is optimized for parameters close to $dE_{\rm dc}/B = 1$, $\hbar\Delta = 0.015B$, and $\Omega/\Delta = 0.9258$ [27] (see below).

We next turn to the dipolar interaction and derive the dressed Born-Openheimer potentials. Each polar molecule is prepared into the internal state $|+\rangle_i$ by an adiabatic switching on of the microwave field. Consequently, the effective interaction potential $V_{\rm eff}(\mathbf{r})$ is determined by the dressed Born-Oppenheimer potential adiabatically connected to the state $|+\rangle_1|+\rangle_2$, see Fig. 2. The competition between the rotational splitting B and the dipole-dipole interaction provides a characteristic length scale $r_{\rm B} = (d^2/B)^{1/3}$. For large interparticle distances $r \gg r_{\rm B}$, the dipole-dipole interaction is weak and does not couple different rotor levels. Consequently, the only relevant coupling appears due to the microwave field bewteen the manifolds with $j = 0$ and $j = 1$. The relevant internal states are then given by the three states $|g\rangle_i = |0,0\rangle_i$, $|e\rangle_i = |1,1\rangle_i$, and $|\bar{e}\rangle_i = |1,-1\rangle_i$. As the microwave field couples each polar molecule with the same phase, the Born-Oppenheimer potentials separate into 6 symmetric and 3 antisymmetric dressed potentials. The effective potential adiabatically connected to the state $|+\rangle_1|+\rangle_2$ is symmetric, and therefore, we can restrict the analysis to the symmeric potentials: a basis is given by the symmetric states $|g,g\rangle$, $|e,g\rangle$, $|e,e\rangle$, $|g,\bar{e}\rangle$, $|e,\bar{e}\rangle$, and $|\bar{e},\bar{e}\rangle$. Within the rotating frame, the Hamiltonian projected onto this subspace reduces to

$$H = \begin{pmatrix} d_g^2\nu & \sqrt{2}\hbar\Omega & 0 & 0 & 0 & 0 \\ \sqrt{2}\hbar\Omega & H_{eg} & \sqrt{2}\hbar\Omega & d_c^2\mu^*/2 & 0 & 0 \\ 0 & \sqrt{2}\hbar\Omega & H_{ee} & 0 & 0 & 0 \\ 0 & d_c^2\mu/2 & 0 & H_{eg} & \hbar\Omega & 0 \\ 0 & 0 & 0 & \hbar\Omega & H_{ee} & 0 \\ 0 & 0 & 0 & 0 & 0 & H_{ee} \end{pmatrix} \quad (3)$$

with the dipole moments $d_g = |\langle g|\mathbf{d}_i|g\rangle|$, $d_e = |\langle e|\mathbf{d}_i|e\rangle|$, and $H_{eg} = (d_e d_g - d_c^2/2)\nu - \hbar\Delta$ and $H_{ee} = d_e^2\nu - 2\hbar\Delta$. The terms $\nu = (1 - 3\cos^2\theta)/r^3$ and $\mu = 3\sin^2\theta e^{2i\phi}/r^3$ describe the spatial dependence of the dipole-dipole interaction, with θ and ϕ being the polar and azimuthal angles of $\mathbf{r}$, respectively. The Born-Oppenheimer potentials then follow from a diagonalization of the Hamiltonian H and are shown in Fig. 2, with the level adiabatically connected to the state $|+\rangle_1|+\rangle_2$ (solid line) giving rise to the effective interaction $V_{\rm eff}(\mathbf{r})$.

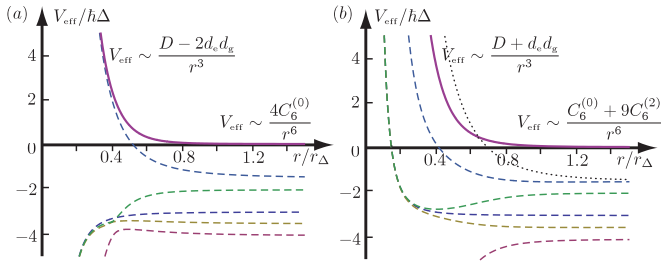

FIG. 2: Born-Oppenheimer potentials in the limit $r \gg r_{\rm B}$: (a) $\theta = 0$ and (b) $\theta = \pi/2$. The effective potential $V_{\rm eff}(\mathbf{r})$ (solid line) is repulsive for all angles θ. The dotted line denotes the antisymmetric level relevant during a three-body collision.

The detuning Δ introduces a new length scale in the

problem $r_\Delta = (d^2/\hbar\Delta)^{1/3} \gg r_B$, e.g., for LiCs with $B/\hbar \approx 5.8$GHz, $r_B \approx 9.2$ nm and $r_\Delta \approx 37.5$nm at $\hbar\Delta = 0.015B$. At large interparticle distances $r > r_\Delta$, the Born-Oppenheimer potentials are well described by perturbation theory in the dipole-dipole interaction. The static electric field gives rise to a dipole moment $d_z = \alpha^2 d_g + \beta^2 d_e$ along the z-axis, while the microwave field drives an additional dipole moment $d_\perp = \sqrt{2}\alpha\beta d_c$ rotating with frequency ω in the x-y plane. The combination of the two dipole forces provides the interaction $V_{\rm eff}(\mathbf{r}) = (d_z^2 - d_\perp^2/2)\left(1 - 3\cos^2\theta\right)/r^3$. A proper choice of the two parameters $E_{\rm dc}d/B$ and Ω/Δ gives $d_z = d_\perp/\sqrt{2}$, providing a cancelation of the leading dipole-dipole interaction [6]. The remaining interaction then follows from second order perturbation theory and provides a van-der-Waals-type repulsion

$$V_{\rm eff}(\mathbf{r}) = \frac{1}{r^6}\left[C_6^{(0)}(1-3\cos^2\theta)^2 + C_6^{(2)}\, 9\sin^4\theta\right] \qquad (4)$$

with

$$\hbar C_6^{(0)} = \frac{\alpha^2\beta^2}{\sqrt{\Delta^2+4\Omega^2}}\left\{\frac{1}{2}\alpha^2\beta^2\left[(d_e-d_g)^2+d_c^2\right]^2 + 2\left[(\alpha^2 d_g+\beta^2 d_e)(d_e-d_g)+\frac{d_c^2}{2}(\beta^2-\alpha^2)\right]^2\right\}, \qquad (5)$$

$$\hbar C_6^{(2)} = \frac{\alpha^4\beta^2 d_c^4}{\Delta+\sqrt{\Delta^2+4\Omega^2}} + \frac{\alpha^2\beta^4 d_c^4}{\Delta+3\sqrt{\Delta^2+4\Omega^2}}.$$

For the optimal values $dE_{\rm dc}/B = 1$ and $\Omega/\Delta = 0.9258$, the van der Waals coefficients take the form $C_6^{(0)} = 0.004\hbar\Delta r_\Delta^6$ and $C_6^{(2)} = 0.005\hbar\Delta r_\Delta^6$. At shorter distances $r_B \ll r < r_\Delta$, the effective interaction reduces to $V_{\rm eff}(\mathbf{r}) = \left(d_c^2 + d_g d_e\left[1-3\cos^2\theta\right]\right)/r^3$ and remains repulsive for all angles θ. Thus it is possible to create purely repulsive interaction with large and adjustable strength.

In order to determine the height of the potential barrier, a detailed analysis including all internal levels is required. Such a procedure is achieved by first deriving Born-Oppenheimer potentials accounting for the coupling of the internal states $|j,m\rangle_i$ by the dipole-dipole interaction. In the second step, the microwave field, which couples these Born-Oppenheimer potentials, is included within a rotating wave approximation. The new dressed levels in the rotating frame are shown in Fig. 3(a). It follows that the height of the shield (solid line) is limited by small avoided crossings. The first crossing (labeled A) appears with the level adiabatically connected to the symmetric state $|1,0;0,0\rangle$ for a relative orientation of the molecules along the z-axis with $\theta \approx 0$, and it limits the barrier height of the shield to $E_{\rm shield} \approx 0.02B$ for the optimal parameters. The radius R_c for the breakdown of the shield is in the range $R_c \sim r_B$, which is still large compared to the distances where additional short range interactions have to be taken into account. This justifies our approach of restricting the analysis to the electric dipole-dipole interaction alone.

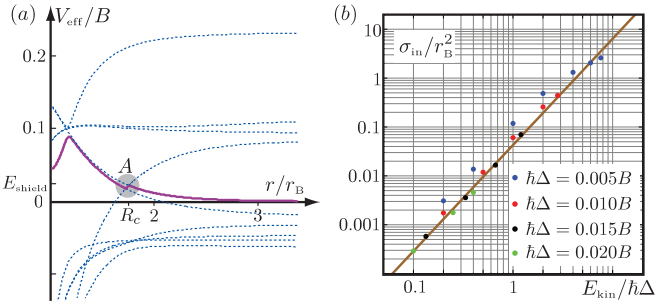

FIG. 3: (a) Born-Oppenheimer potentials for $\hbar\Delta = 0.015B$ at $\theta = 0$. The plot shows all potentials (dashed) accessible via single-photon transitions from the state (solid) adiabatically connected to $|+\rangle_1|+\rangle_2$. The first crossing limiting the height of the shield appears in the region A for angles $\theta \approx 0$. (b) Inelastic cross section $\sigma_{\rm in}$ due to diabatic crossings for different detunings Δ as a function of incoming kinetic energy $E_{\rm kin}$.

Next, we analyze the validity of the Born-Oppenheimer approximation and study the influence of the kinetic energy coupling different dressed potentials during a collision. The influence of the kinetic energy is determined by the dimensionless parameter $\gamma = d^2 m/\hbar^2 r_B$. For $\gamma \gg 1$, we can apply semiclassical theory; this condition is well satisfied for typical polar molecules like LiCs with $\gamma \approx 6900$. For a collision whose relative kinetic energy $E_{\rm kin}$ is below the shield barrier, the processes giving rise to inelastic loss are (i) diabatic crossing between different Born-Oppenheimer levels and (ii) quantum mechanical tunneling through the barrier. We start by studying the diabatic transitions first: the inelastic cross section within semi-classical approximation is computed by first determining the classical trajectory $r_{\rm cl}(t)$ of a collision with a fixed impact parameter. To determine the Landau-Zener diabatic crossings, we solve the full Schrödinger equation for the internal structure using the given relative motion $r_{\rm cl}(t)$. The loss propability is then determined by the depletion of the adiabatic Born-Oppenheimer level at the classical turning point. Averaging over different impact parameters and angles of approach provides the inelastic cross section $\sigma_{\rm in}$ due to diabatic transitions. The main contribution comes from the Born-Oppenheimer level closely approaching the effective potential close to $\theta \approx 0$, see Figs. 2(a) and 3(a). Note that the standard Landau-Zener tunneling expression can not be applied here as the levels have no real crossing. The inelastic cross section for different Δ and $E_{\rm kin}$ is shown in Fig. 3(b). We find an algebraic behavior of the inelastic cross section with $\sigma_{\rm in} = \rho(E_{\rm kin}/\hbar\Delta)^\kappa r_B^2$ with $\kappa \approx 2.2$ and $\rho = 0.043$ at $\hbar\Delta = 0.015B$, solid line in Fig. 3(b). The loss rate $1/\tau_{\rm in}$ during a two-particle collisions reduces to

$$\frac{1}{\tau_{\rm in}} \approx 11\frac{B\, n r_B^3}{\hbar}\left(\frac{T}{B}\right)^{\kappa+1/2}, \qquad (6)$$

where we have replaced the collision energy with the temperature T of the gas. Consequently, for LiCs at characteristic densities $n \sim 10^{13}\,\mathrm{cm}^{-3}$, the lifetime is several seconds even for $T \sim 1\mathrm{mK} < \hbar\Delta$.

The second scenario for an inelastic collision is quantum mechanical tunneling through the barrier. For $\gamma \gg 1$ such tunneling processes are strongly suppressed and can be studied using semiclassical techniques like WKB. The tunneling propability during a single collision is then given by the Eucledian action for the trajectory C starting at the classical turning point R_0 and ending at the inner distance R_c, where the shield starts to break down;

$$P_{\mathrm{WKB}} = \exp\left(-2\int_C ds\,\sqrt{m\left[V_{\mathrm{eff}}(\mathbf{r}) - E_{\mathrm{kin}}\right]}/\hbar\right). \quad (7)$$

Thus, the highest tunneling probability is obtained along the collision axis with the lowest shield barrier, see discussion above. Then the characteritic scale for the tunneling amplitude at low incoming kinetic energies ($E_{\mathrm{kin}} \ll \hbar\Delta$) is given by $P_{\mathrm{WKB}} = \exp(-c\sqrt{\gamma})$, where the numerical constant for $\hbar\Delta/B = 0.015$ takes the form $c \approx 0.32$. Consequently, for LiCs the tunneling is strongly suppressed and can be safely ignored at low kinetic energies $E_{\mathrm{kin}} < \hbar\Delta$.

It is important to note that the qualitative behavior of the shield remains robust during a three-body collision. The main modification to the shield is that the antisymmetric levels can open up an avoided crossing, as the parity symmetry present in the two-particle problem is broken for three particles. The relevant antisymmetric level is shown by a dotted line in Fig. 2(b). As this crossing appears at energies $\sim\hbar\Delta$, it does not modify the validity of the above discussion for incoming kinetic energies $E_{\mathrm{kin}} < \hbar\Delta$. Thus, the shield prevents three particles from approaching each other on distances, where the formation of bound states can take place, and three-body losses are therefore effectively quenched.

To summarize, we showed that properly adjusted continous wave microwave and DC electric fields can create a repulsive shield resulting in large suppression of inelastic collisions. We now describe possible avenues opened by this work. For efficient evaporative cooling, it is important that elastic collisions allow for fast thermalization. The elastic scattering cross section at low collisional energies is determined by the s-wave scattering length. In the present situation, the scattering length can be estimated via a partial wave expansion [25]: the dominant part is determined by the isotropic part of the effective interaction potential $V_{\mathrm{eff}}^{(0)}(r) = C_6/r^6$. The s-wave scattering length then reduces to $a_s \approx \left(C_6 m/\hbar^2\right)^{1/4} \sim r_\Delta \left(d^2 m/\hbar^2 r_\Delta\right)^{1/4}$. For LiCs at the optimal detuning $\hbar\Delta = 0.015B$, it follows that $a_s \approx 66\,\mathrm{nm}$ yielding a large elastic cross section σ_{el} with suppressed losses providing an ideal system for evaporative cooling, e.g. $\sigma_{\mathrm{el}}/\sigma_{\mathrm{in}} \sim 10^6$ for σ_{in} at $T = 1$ mK and $\sigma_{\mathrm{el}} = 8\pi a_s^2$.

Another application is the creation of three-dimensional crystalline phases. The many-body Hamiltonian for a gas of ultracold polar molecules reduces to

$$H = \sum_i \frac{\mathbf{p}_i^2}{2m} + \frac{1}{2}\sum_{i\neq j} \frac{C_6}{|\mathbf{r}_i - \mathbf{r}_j|^6}, \quad (8)$$

where we neglect additional terms due to the anisotropy of the effective potential and due to three-body interactions discussed in [6]. In analogy to the appearance of crystalline phases for polar molecules confined to two dimensions [26], for strong repulsive van der Waals interactions, the system will undergo a phase transition from a liquid phase to a solid phase. The dimensionless parameter controlling the transition takes the form $\lambda = C_6 m/\hbar^2 a^4$ with $n = 1/a^3$ the particle density: for weak interactions $\lambda \ll 1$ the ground state is in a liquid phase, while for strong interactions with $\lambda \gg 1$ the system is characterized by a solid phase with broken translational symmetry. Consequently, this opens up a way towards the creation of three-dimensional crystalline structures with ultracold molecular gases.

We acknowledge discussions with J. Doyle and W. Campbell. The work was supported by the European Science Foundation with EuroQUAM program, FWF, NSF, ARO, MURI, and DARPA.

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